

WHAT IS CLAIMED IS:

1. A lead-free glass flux suitable for coating onto a glazed surface, comprising the following two glass compositions:

5 (a) a glass composition which is essentially free from lead and which comprises, in mass percent, SiO<sub>2</sub>: 45 to 60%, Al<sub>2</sub>O<sub>3</sub>: 5 to 20%, B<sub>2</sub>O<sub>3</sub>: 15 to 35%, and one or more alkali metal oxides: 5 to 10%, provided that Li<sub>2</sub>O is contained in an amount of 2% or more, with the proviso that the total amount of said oxides is 90% or more of the total mass of the composition; and

10 (b) a glass composition which is essentially free from lead and which comprises, in mass percent, SiO<sub>2</sub>: 45 to 60%, Al<sub>2</sub>O<sub>3</sub>: 5 to 20%, B<sub>2</sub>O<sub>3</sub>: 5 to 20%, at least one of MgO, CaO, ZnO: 5 to 20% in total, and one or more alkali metal oxides: 10 to 15%, provided that Li<sub>2</sub>O is contained in an amount of 2% or more, with the proviso that the total amount of said oxides is 90% or more of the total mass of the composition,

15 wherein the glass compositions (a) and (b) are mixed such that the coefficient of thermal expansion of a glass molded product formed from the flux is  $6.5 \times 10^{-6} \text{K}^{-1}$  or less.

20 2. The glass flux according to claim 1, wherein the coefficient of thermal expansion of the glass composition (a) is in the range of  $5.0 \times 10^{-6} \text{K}^{-1}$  to  $6.5 \times 10^{-6} \text{K}^{-1}$  and the coefficient of thermal expansion of the glass composition (b) is in the range of  $7.0 \times 10^{-6} \text{K}^{-1}$  to  $9.0 \times 10^{-6} \text{K}^{-1}$ .

25 3. The glass flux according to claim 1, wherein the softening point of each of the glass composition (a) and the glass composition (b) is in the range of 500 to 600°C.

30 4. The glass flux according to claim 1, wherein the mixing ratio of the glass composition (a) and the glass composition (b) in a mass ratio is 5 to 25 parts of the glass composition (b) to 75 to 95 parts of the glass composition (a), provided that the total of (a) and (b) is 100 parts.

5. The glass flux according to claim 1, wherein the glass composition (a) and the glass composition (b) that are mixed are each prepared in powdered form.
6. A composition suitable for the decoration of ceramic materials, comprising:  
5 a lead-free glass flux as claimed in claim 1 and at least one pigment.
7. The composition according to claim 6, further comprising at least one low expansion ceramic raw material in a content of 10% or less of the total mass of the composition.
- 10 8. The composition according to claim 6, wherein the composition is substantially composed of, in mass percent, 70 to 95% of said lead-free glass flux and 5 to 30% of at least one pigment.
- 15 9. A method for making a composition suitable for the decoration of ceramic materials, comprising the steps of:  
separately preparing:  
(a) a glass composition which is essentially free from lead, is composed mainly of  $\text{SiO}_2$  and comprises  $\text{Al}_2\text{O}_3$ ,  $\text{B}_2\text{O}_3$  and  $\text{Li}_2\text{O}$  as essential ingredients, whose coefficient  
20 of thermal expansion is in the range of  $5.0 \times 10^{-6}\text{K}^{-1}$  to  $6.5 \times 10^{-6}\text{K}^{-1}$ ;  
(b) a glass composition which is essentially free from lead, is composed mainly of  $\text{SiO}_2$  and comprises  $\text{Al}_2\text{O}_3$ ,  $\text{B}_2\text{O}_3$ ,  $\text{Li}_2\text{O}$  and at least one of  $\text{MgO}$ ,  $\text{CaO}$  and  $\text{ZnO}$ , as essential ingredients, whose coefficient of thermal expansion is in the range of  $7.0 \times 10^{-6}\text{K}^{-1}$  to  $9.0 \times 10^{-6}\text{K}^{-1}$ ;  
25 (c) at least one pigment; and  
mixing the prepared glass composition (a), glass composition (b) and pigment (c) such that a decorating material having a coefficient of thermal expansion of  $7.0 \times 10^{-6}\text{K}^{-1}$  or less is obtained as a final product.
- 30 10. The method according to claim 9, wherein the softening point of each of the glass composition (a) and the glass composition (b) is in the range of 500 to 600°C.

11. The method according to claim 9, wherein

the glass composition (a) comprises, in mass percent, SiO<sub>2</sub>: 45 to 60%, Al<sub>2</sub>O<sub>3</sub>: 5 to 20%, B<sub>2</sub>O<sub>3</sub>: 15 to 35%, and one or more alkali metal oxides: 5 to 10%, provided that  
5 Li<sub>2</sub>O is contained in an amount of 2% or more, with the proviso that the total amount of said oxides is 90% or more of the total mass of the composition, and wherein

the glass composition (b) comprises, in mass percent, SiO<sub>2</sub>: 45 to 60%, Al<sub>2</sub>O<sub>3</sub>: 5 to 20%, B<sub>2</sub>O<sub>3</sub>: 5 to 20%, at least one of MgO, CaO, ZnO: 5 to 20% in total, and one or more alkali metal oxides: 10 to 15%, provided that Li<sub>2</sub>O is contained in an amount of  
10 2% or more, with the proviso that the total amount of said oxides is 90% or more of the total mass of the composition.

12. The method according to claim 9, wherein at least one low expansion ceramic raw material is further mixed in an amount of 10% or less of the total mass of the  
15 composition in the mixing step.

13. The method according to claim 9, wherein the glass composition (a) and the glass composition (b) are mixed in the ratio on a mass basis of 5 to 25 parts of the glass composition (b) to 75 to 95 parts of the glass composition (a), provided that the total of  
20 (a) and (b) is 100 parts, thereby obtaining a mixture which comprises, in mass percent, 70 to 95% of the total of the glass composition (a) and the glass composition (b) and 5 to 30% of the pigment (c), provided that the mixture obtained as a final product is substantially composed of the glass composition (a), the glass composition (b) and the  
25 pigment (c).

14. A ceramic article which has been decorated using a composition as claimed in claim 6.